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13. ABSTRACT (Maximum 200 words)

This project addressed several major questions concerning the coherence of electron spins in Si-based structures. Decoherence of the qubits is a fundamental limitation of any quantum computing device. Our goal is to understand the origin of spin decoherence for both free and bound electrons in Si. We have found that electrons bound to donors can have extremely long coherence times in isotopically enriched 28Si, with the ratio of their coherence time to their oscillation frequency being nearly 109. On the other hand we find that free electrons decohere at least 4 orders of magnitude faster than the bound electrons. In addition to electrons in pristine silicon crystals, we have investigated spin coherence for ion-implanted donors and find that their decoherence is larger, but with coherence times of > 1ms. Etching the oxide off of the silicon surface reduces the decoherence by about a factor of 2, pointing to the need to carefully control defects in all parts of the structures.

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Final Progress Report
Electron Spin Decoherence in Si-Based Structures

This project began with 4 major goals:
1. Understand the origin of the decoherence (or phase memory) of electron spins bound to donors in Si.
2. Understand the origin of the decoherence of free electron spins in Si quantum wells embedded in SiGe
3. Determine how confinement (quantum dots and wires) affects the coherence of electron spins in Si
4. Determine the efficacy of optical injection for generating spin-aligned electrons in Si.

At the time this program was begun the limits on the electron spin coherence time in Si were not known. The spin-flip time (T_1) for electrons bound to donors was known to be long – minutes to hours at ~1K, but there had only been two reports of the phase memory or spin coherence time (which I will call T_2 here). The last T_2 measurements had been done in the early 1970's, and the only measurement of T_2 for isotopically enriched ^{28}Si dated from 1958 (and were only the second measurement of electron spin echoes in any system). Before this program began we had made preliminary spin-echo measurements of T_2 in natural Si, and had found that the echo decays were nonexponential, not even monotonic. In addition, no pulsed Electron Spin Resonance (ESR) measurements had been made of two-dimensional (2D) electrons in Si. Continuous wave (cw) ESR measurements of 2D electrons gave quite narrow lines, requiring T_1 and T_2 to be at least a few microseconds. No ESR measurements had been done at that time on Si quantum dots, either. Our original plan was to measure spin coherence, and we suspected that the 2D electrons would have a T_2 comparable to that of the donor electrons (at least 100 μs), and then to demonstrate a charge-coupled device (CCD) which would transport spin-coherent electrons. In the course of this work we found that this supposition was erroneous, and that both T_1 and T_2 are of the order of a microsecond for 2D electrons, and we did not pursue the CCD structure. The optical injection was to be used for the CCD, and thus we did not pursue that line of work, either.

The two major results of this program have been the measurement of T_2 for electrons bound to donors in Si with the discovery that their coherence time can be at least 2 orders of magnitude longer than was seen in the 1958 experiments, and the measurement of T_1 and T_2 for high-mobility 2D electrons and the result that their spin relaxation is about 4 orders of magnitude faster than that of the donor-bound electrons. In the case of electrons bound to phosphorus donors in isotopically enriched ^{28}Si we have shown that if we extrapolate to isolated electrons (so that they do not interact with one another by their magnetic moments) their spin coherence time is ~60 ms at about 7K. This can be looked at as a Q-factor for the resonance of almost 10^9. Since these times are long, small perturbations from the environment can mask the true coherence time. In our experiments (and probably the 1958 ones) we found that fluctuating magnetic fields in the lab were accelerating the apparent echo decay. After dealing with the field fluctuations, we found conventional spin echo decays of up to ~ 4 ms (while the 1958 results were 0.52 ms). However, from the dependence of T_2 on the doping density is was

clear that this 4 ms was coming from an aspect of the magnetic dipole-dipole coupling between the electrons, which is termed instantaneous diffusion. An established technique for quantifying the effect of instantaneous diffusion is to perform a usual 2-pulse Hahn echo experiment, but vary the strength of the second, refocusing, pulse. For small pulses the dipole-dipole term is reduced, and the echo decay becomes longer. That is what we found experimentally. For example, if the second pulse caused a 45° rotation (rather than the usual 180°) the echo decay time increased to about 15 ms. Extrapolating to a refocusing pulse of zero amplitude gives the T_2 = 60 ms result for an isolated donor electron spin.). This work is [1] in the publication list below.

As a function of temperature we found that $T_2 \approx T_1$ for temperatures down to about 7K, which is where we measured these long coherence times. At lower temperature T_1 becomes longer, but T_2 does not appear to increase. This saturation of T_2 may be evidence for further magnetic field noise or other noise from the ESR spectrometer. It is also likely that the coherence is being limited by the residual ^{29}Si in our samples (~800 ppm as measured by SIMS for us by Prof. Kohei Itoh of Keio University in Japan.

The second major accomplishment has been the measurement of T_1 and T_2 for 2D electrons in high-mobility Si quantum wells in Si/SiGe heterostructures. These measurements were the first reported pulsed-ESR measurements of 2D electrons. We found a maximum T_2 of ~ 3 μs, with $T_1 \approx T_2/2$. The fact that $T_2 > T_1$ is unusual for spins, and indicates that the relaxation is dominated by a spin-flip process (a purely longitudinal relaxation process). It appears that the relaxation can be understood as arising from the Rashba effect – a spin-orbit interaction term which is zero in the bulk of the Si, by inversion symmetry, but the symmetry is broken by the quantum well. The asymmetry in the quantum well can come from an electric field (the modulation doping) and from differences between the two interfaces. This Rashba effect leads to an effective magnetic field which is directed perpendicular to the electron's momentum, and thus fluctuates as the electron undergoes momentum scattering. The effective field lies in the plane of the 2D electrons, and thus for a perpendicular magnetic field it causes purely longitudinal spin relaxation (the fluctuating effective field is always perpendicular to the applied field). In this picture a more symmetric quantum well would give a longer T_1 and T_2, but we have studied several samples and seen little difference. These results appear in [2].

The measurements of spin relaxation give us a handle on what to expect for spins in quantum dots and quantum wires, in that we have a measurement of the spin-orbit interaction effects. The simplest case, conceptually, would be to make structures small enough that electrons could be frozen into a ground spatial state. In that case the quantum dot would be expected to act like a donor, and a spin in a quantum wire will pick up a phase which is uniquely determined by the distance along the wire it has traveled. However, recent work on the valley splitting for 2D electrons at the University of Wisconsin-Madison has shown that the two remaining valleys in a Si quantum well are nearly degenerate. Thus, it is not clear whether making a small dot is sufficient to ensure that the lowest state in the dot is only 2-fold (spin) degenerate. Wisconsin found that the valley splitting was small, and it is not known exactly what controls the splitting. It is possible that some of the decoherence we observed for 2D electrons came from

intervalley scattering effects, rather than just a Rashba term, but that will require further work to sort out.

In other work we have collaborated with the Prof. Mark Eriksson's group at University of Wisconsin-Madison to measure decoherence in quantum dots. The dots have been fabricated at Wisconsin, and we have made the pulsed-ESR measurements. During this project we only had time to study rather large dots – of the order of 1 μm, and larger. At this size the spin relaxation was essentially that same as what we measured in the 2D electron samples.

We also began a collaboration with Dr. Thomas Schenkel's group at Lawrence Berkeley Lab on spin coherence of electrons bound to implanted donors. With implantation the donors can be placed very near to the silicon surface, and the process will also generate defects which may not be completely annealed. The effect of the surface and other defects has not been previously measured. We have not yet determined whether bulk Si defects are affecting T_2 of these donors, but we can still observe coherence times of 1 - 2 ms. We do see that the state of the Si surface plays a role. After etching the oxide off in HF, which should leave the Si surface partially hydrogen terminated (not well-terminated because the silicon was (100) oriented and after etching it was not possible to hermetically seal the sample) the spin coherence approximately doubled. Apparently the donor spins were either interacting through their magnetic moment with other paramagnetic centers in the oxide or at the Si/SiO_2 interface, or there were fluctuations in the charge state of traps which decohered the donor electron spins through a Stark shift of the spin resonance frequency. A preliminary report on this work has been submitted to Applied Physics Letters [8].

We have also collaborated with a group at Oxford University to study spin coherence of nitrogen-doped C_{60} ($N@C_{60}$). That project is primarily supported by Princeton and the NSF, but there has been some overlap with this ARO project and the PI has spent some time working on it, and thus we have acknowledged ARO support in the papers which have been produced. Some of the results are particular to $N@C_{60}$ (for example T_2 measurements[3], and measurements of some peculiarities in the echo decays from this molecule[4]) This collaboration has also led to some results which are of a broader interest to quantum information processing. We have developed methods to quantify certain errors, in particular phase errors in single-qubit gates (microwave pulses for ESR)[5]. We have also shown that certain pulse sequences which have been developed for nuclear magnetic resonance may also be used to produce highly accurate electron spin rotations[6]. Finally, we have controlled the nitrogen nuclear spin through the spin of the electron, and demonstrated bang-bang decoupling of the nuclear spin from noise[7].

The refereed journal publications which acknowledged support from this ARO project are:

1. A.M. Tyryshkin, S.A. Lyon, A.V. Astashkin, and A.M. Raitsimring, "Electron Spin-Relaxation Times of Phosphorus Donors in Silicon," Phys. Rev. B **68**, 193207 (2003).

2. A.M. Tyryshkin, S.A. Lyon, W. Jantsch, and F. Schäffler, “Spin Manipulation of Free 2-Dimensional Electrons in Si/SiGe Quantum Wells,” Phys. Rev. Lett. **94**, 126802, (2005).
3. John J. L. Morton, Alexei M. Tyryshkin, Arzhang Ardavan, Kyriakos Porfyrakis, S.A. Lyon, and G. Andrew D. Briggs “Electron spin relaxation of N@C_{60} in CS_2” accepted for publication in J. Chem. Phys.
4. J.J. Morton, A.M. Tyryshkin, A. Ardavan, K. Porfyrakis, S.A. Lyon, and G.A.D. Briggs, “A New Mechanism for Electron Spin Echo Envelope Modulation,” J. of Chem. Phys. **122**, 174504 (2005).
5. J.J. Morton, A.M. Tyryshkin, A. Ardavan, K. Porfyrakis, S.A. Lyon, and G.A.D. Briggs, “Measuring Errors in Single-Qubit Rotations by Pulsed Electron Paramagnetic Resonance,” Phys. Rev. A **71**, 012332, (2005).
6. J.J. Morton, A.M. Tyryshkin, A. Ardavan, K. Porfyrakis, S.A. Lyon, and G.A.D. Briggs, “High Fidelity Single Qubit Operations Using Pulsed Electron Paramagnetic Resonance,” accepted for publication in Phys. Rev. Lett., available as quant-ph/0502119.
7. John J. L. Morton,_ Alexei M. Tyryshkin, Arzhang Ardavan, Simon Benjamin, Kyriakos Porfyrakis, S. A. Lyon, and G. Andrew D. Briggs, “Bang-bang control of fullerene qubits using fast Berry phase gates”, accepted for publication in Nat. Phys.

Papers submitted but not yet accepted for publication in refereed journals:

8. T. Schenkel, A.M. Tyryshkin, R. de Sousa, K.B. Whaley, J. Bokor, J.A. Liddle, A. Persaud, J. Shangkuan, I. Chakarov, and S.A. Lyon, “Electrical activation and electron spin coherence of ultra-low dose antimony implants in silicon”.

Conference presentations:

9. Alexei M. Tyryshkin, S.A. Lyon, Wolfgang Jantsch, and Friedrich Schäffler, “Manipulation of Free 2D Electron Spins by Pulsed ESR”, MRS Fall Meeting, Boston (2002).
10. Alexei M. Tyryshkin, S.A. Lyon, Wolfgang Jantsch, and Friedrich Schäffler, “Spin Relaxation Times of Conduction Electrons in Si/SiGe Quantum Wells”, 25th International EPR Symposium, Denver (2002).
11. Alexei M. Tyryshkin, Stephen A. Lyon, Andrei V. Astashkin, Arnold M. Raitsimring, “Mechanisms of Electron Spin Relaxation of Phosphorus Donors in Silicon,” 25th International EPR Symposium, Denver (2002).
12. S.A. Lyon, “Spin Relaxation in Phosphorus Doped Si”, (invited) APS March Meeting, Montreal (2002).
13. A.M. Tyryshkin, “Relaxation time of electron spins in Si structures,” (invited) 34th Winter Colloquium on the Physics of Quantum Electronics, Snowbird (2004).
14. A. M. Tyryshkin, S.A. Lyon, A. V. Astashkin, A. M. Raitsimring, “Exceptionally Long Electron Spin Relaxation Times of Phosphorus Donors in Silicon,” 27th International EPR Symposium, Denver (2004).
15. A.M. Tyryshkin, J.J.L. Morton, A. Ardavan, K. Porfyrakis, S.A. Lyon, G.A.D. Briggs, “Electron Spin Echo Envelope Modulation caused by isotropic hyperfine coupling in liquid solutions,” 27th International EPR Symposium, Denver (2004).

16. A.M. Tyryshkin, J.J.L. Morton, A. Ardavan, K. Porfyrakis, S.A. Lyon, G.A.D. Briggs, "Electron spin relaxation times of endohedral fullerene N@C_{60} in liquid solutions," 27th International EPR Symposium, Denver (2004).
17. A. M. Tyryshkin, S. A. Lyon, W. Jantsch, and F. Schäffler, "Pulsed Electrically-Detected Magnetic Resonance of 2D Electrons in a Si/SiGe Quantum Well," APS March Meeting, Montreal (2005).